The molecule is nearly planar and the bond lengths and bond angles are normal However, there is no dimerization of carboxyl groups, rather the molecules within the same layer are linked through hydrogen bond (N. .H...O) of 2·84 Å between the nitrogen atom of one molecule to the carbonyl oxygen atom of the other Apart from this, there is an intramolecular hydrogen bond of 2·43 Å as in maleic acid structure Further work to refine the structure is in progress

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Theory of Brillouin scattering in an absorbing thin film

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Observation of Brillouin scattering from thin films has been reported by Sandercock (1972) He derived the spectra associated with thin films and bulk specimens Recently, Dervisch & Loudon (1976) have derived Lorentzian and skewed spectral lineshapes for a semi-infinite specimen In the present paper we give a comprehensive theory of Brillouin scattering in reflection from an absorbing thin film

Consider a thin film of thickness d occupying the space $-d < z < 0$, and boundaries being in the xy plane We treat the incident radiation as having its plane of polarisation normal to the plane of incidence (s-waves). If the crystal excitations are of wavevector q and frequency w, then their coupling to incident radiation of wavevector k_L and frequency w_L generates a Stokes polarisation *inside* the crystal of the form

$$P_s e^{i(kz - W_s t)} \qquad \ldots (1)$$

where

$$k = k_L - q \qquad \ldots (2)$$

$$w_s = w_L - w \qquad \ldots (3)$$

The Stokes polarisation generates electromagnetic waves in region I ($z > 0$) and region II (inside the specimen). A formal solution also shows existence of scattered waves in region III ($z < -d$), and this is the forward scattered component We are, however, interested in scattering in reflection, a technique

first employed by Russel (1965) and is very useful in studying absorbing materials By solving Maxwell's equations and applying the usual boundary conditions, which are, the tangential electric field and normal displacement vector are continuous, we obtain the electric field in the space $z > 0$ as

$$E_{1s} = \left\{ \frac{1}{[k^2 - k_{2s}^2]} + \frac{[k_{1s}-k][e^{-i(k_{2s}+k)d} - 1]}{[k^2-k_{2s}^2][k_{1s}-k_{2s}][1-e^{-2ik_{2s}d}]} + \frac{[k_{1s}-k][e^{i(k_{2s}-k)d} - 1]}{[k^2-k_{2s}^2][k_{1s}+k_{2s}][1-e^{2ik_{2s}d}]} \right\} \frac{w_s^2 P_s e^{i(k_{1s}z - w_s t)}}{\epsilon_0 c^2} \quad \ldots (4)$$

and the electric field inside $(-d < z < 0)$ is

$$E_{2s} = \left\{ \frac{e^{ikz}}{[k^2-k_{2s}^2]} + \frac{[k_{1s}-k][e^{-i(k_{2s}+k)d} - 1]e^{ik_{2s}z}}{[k^2-k_{2s}^2][k_{1s}-k_{2s}][1-e^{-2ik_{2s}d}]} + \frac{[k_{1s}-k][e^{i(k_{2s}-k)d} - 1]e^{-ik_{2s}z}}{[k^2-k_{2s}^2][k_{1s}+k_{2s}][1-e^{2ik_{2s}d}]} \right\} \frac{w_s^2 P_s e^{-iw_s t}}{\epsilon_0 c^2} \quad \ldots (5)$$

where k_{1s} and k_{2s} are wavevectors of the scattered light in regions I and II respectively.

$$k_{1s}^2 = \frac{w_s^2}{c^2} \epsilon_1$$

$$k_{2s}^2 = \frac{w_s^2}{c^2} \epsilon_2(w_s) \quad \ldots (6)$$

where ϵ_1 is a frequency independent dielectric constant for region I and $\epsilon_2(w_s)$ is a frequency dependent dielectric function for region II. Eqs (4) and (5) are exact In these equations the second and third terms contain factors depending on the size of the specimen Such dependence have been mentioned by Dresselhaus & Pine (1975) It can be mentioned that eqs. (4) and (5) are in fact modifications of results of Dervisch & Loudon (1976) when effects of two boundaries are considered

We are interested in the scattered light observed in reflection, and so we shall consider eq (4) There are three terms, and each will contribute to the overall observable effects, but it can be noted that the first term does not have any size dependence The second term will show the dominant size effects.

The final term is negligible in comparison with the second term, since it has the denominator $(k_{1s}+k_{2s})$ which is very large ($\sim 10^6$ cm^{-1}). Such sums appearing in denominators have been discussed in Raman scattering theories of thin films by Chen *et al* (1975) and Nkoma (1975). Thus the dominant term for size effects is

$$E_{1s} \approx \left\{ \frac{(k_{1s}-k_L+q)[e^{-i(k_L+k_{2s}-q)d}-1]}{q(k_{1s}-k_{2s})(k_L+k_{2s}-q)[e^{-2ik_{2s}d}-1]} \right\} \frac{w_s^2 P_s e^{i(s_1 z-w_s t)}}{\epsilon_0 c^2} \quad \ldots (8)$$

The wavevectors k_{1s} and q are real, while k_L and k_{2s} are complex

$$k_L = k_L' + ik_L'' \quad \ldots (9)$$

$$k_{2s} = k'_{2s} + ik''_{2s} \quad \ldots (10)$$

If we neglect any differences in the crystal optical properties at the frequencies w_L and w_s then

$$k'_L = k'_{2s} = \frac{w_s}{c} n_1 = k_0 n_1 \quad \ldots (11)$$

$$k''_L = k''_{2s} = \frac{w_s}{c} n_2 = k_0 n_2 \quad \ldots (12)$$

where n_1 and n_2 are the refractive index and extinction coefficient respectively

The cycle average intensity of the scattered light is obtained as

$$I_s = \frac{g_s w_s^2 \left\{ 1 - \dfrac{\cos(2n_1 k_0 - q)d}{\cosh(2n_2 k_0 d)} \right\}}{4c^2 \{(2n_1 k_0 - q)^2 + 4(n_2 k_0)^2\} \left\{ 1 - \dfrac{\cos(2n_1 k_0 d)}{\cosh(2n_2 k_0 d)} \right\}} \quad \ldots (13)$$

where

$$g_s = \frac{w_s^2 |P_s|^2 \{(k_{1s} - n_1 k_0 + q)^2 + (n_2 k_0)^2\}}{8\epsilon_0 q^2 \{(k_{1s} - n_1 k_0)^2 + (n_2 k_0)^2\}} \quad \ldots (14)$$

Eq (13) is the one which contains the behaviour of the scattered spectrum from an absorbing thin film. There are three cases of practical interest.

First, consider the case of a non-absorbing thin film, whence $n_2 = 0$. The intensity becomes

$$\frac{f_s w_s^2}{(n_1 k_0 c)^2} \frac{\sin^2[(2n_1 k_0 - q)d/2]}{[(2n_1 k_0 - q)d/2]^2} \quad \ldots (15)$$

where

$$f_s = \lim_{n_2 \to 0} g_s$$

and we have used $\sin n_1 k_0 d \approx n_1 k_0 d$

The spectrum dependence obtained in eq. (15) has been observed by Sandercock (1972).

Secondly, consider a semi-infinite absorbing specimen, whence the limit $d \to \infty$ and $n_2 \neq 0$ In this case the intensity becomes

$$\frac{g_s w_s^2}{4c^2\{(2n_1k_0-q)^2+4(n_2k_0)^2\}}. \quad \ldots (16)$$

This is the case discussed by Dervisch & Loudon (1976), and Dresselhaus & Pine (1975) amongst others However, these authors and others have discussed cases where the line shape in eq (16) is modified to a skew line shape by considering the effect of an equal but opposite Stokes polarisation

Thirdly, consider a non-absorbing medium ($n_2 = 0$) and thickness $d \to \infty$. The observed intensity becomes

$$\frac{w_s^4 |P_s|^2 (k_{1s}+n_1k_0}{8c_0c^3q^2(n_1k_0)^2(k_{1s}-n_1k_0)^2} \pi\delta(2n_1k_0-q) \quad .. (17)$$

The δ-function behaviour in the present case leads to the normal Brillouin shift $\Omega_0 = 2n_1k_0v$, where v is the velocity of sound

In conclusion, we have derived eq. (13) which describes the scattered light spectrum from a thin film The results in the cases of a transparent thin film, an absorbing and non-absorbing semi-infinite specimen have been obtained as special cases The special cases have previously been discussed separately by several authors (Dervisch & Loudon 1976, Sandercock 1972).

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